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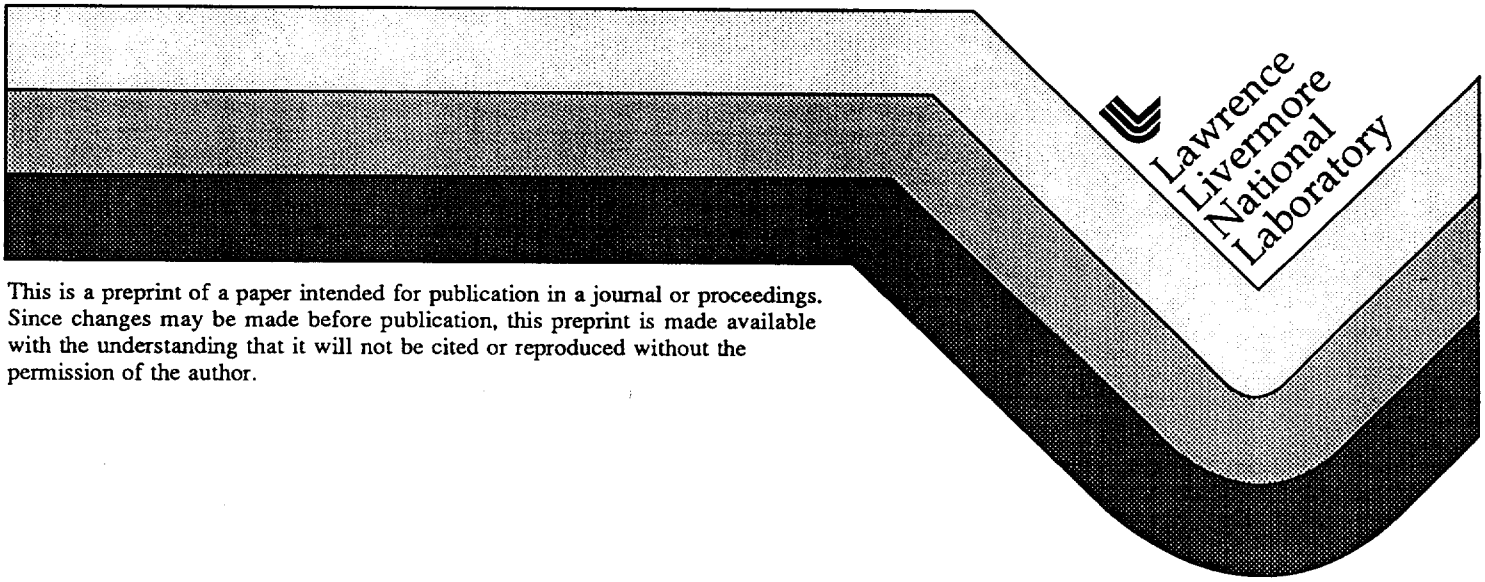

IMPACT OF HIGH SPEED CIVIL TRANSPORTS ON STRATOSPHERIC OZONE: A 2-D MODEL INVESTIGATION

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Abstract

This study investigates the effect on stratospheric ozone from a fleet of proposed High Speed Civil Transports (HSCTs). The new LLNL 2-D operator-split chemical-radiative-transport model of the troposphere and stratosphere is used for this HSCT investigation. This model is integrated in a diurnal manner, using an implicit numerical solver. Therefore, rate coefficients are not modified by any sort of diurnal average factor. This model also does not make any assumptions on lumping of chemical species into families. Comparisons to previous model-derived HSCT assessment of ozone change are made, both to the previous LLNL 2-D model and to other models from the international assessment modeling community. The sensitivity to the NO_x emission index and sulfate surface area density is also explored.

1. INTRODUCTION

The aviation community is investigating the possibility of developing, marketing, and producing a fleet of High Speed Civil Transports or HSCTs [1]. These aircraft are designed to cruise at Mach 2.4 with a range of 5000 to 6500 nautical miles. The primary market for this new fleet of supersonic passenger aircraft will be the Atlantic and Pacific flight corridors, decreasing the average subsonic travel time by over a factor of two. This HSCT fleet will cruise primarily in the lower stratosphere, within an ozone rich region. Effluents from this proposed fleet will predominately be emitted within the Northern Hemisphere, mid-latitude, lower stratosphere flight corridor. Emissions of trace constituents from HSCTs engines are produced from: 1) direct combustion of the kerosene based fuel components forming H_2O, CO_2, CO, CH_4, non-methane hydrocarbons, and soot; 2) impurities in the fuel (e.g., sulfur, forming SO_2); and 3) high temperature processes, breaking down atmospheric nitrogen forming NO_x ($NO+NO_2$).

Currently, the NASA Atmospheric Effects of Stratospheric Aircraft Program (AEAP) is investigating the potential impact of the above mentioned HSCT fleet emissions on ozone. Within the direction of the AEAP, a group of models taken from the international community have completed an assessment of such a fleet [1]. In *Figure 1*, a example of the spread in model results are shown. Here, for March, at 45°N latitude, the change in local ozone concentration is shown for a 500 aircraft Mach 2.4 fleet, assuming an emission index (EI) of NO_x of 5 (grams as NO_2 per kg of fuel). This proposed fleet is composed of both HSCTs and next generation subsonic aircraft, which are needed to meet market projections in the year 2015. The reference atmosphere is assumed to be a projected 2015 fleet of subsonic aircraft, assuming HSCTs are not built. In the lower stratosphere, the spread in model-derived ozone change was between +5% (CAMED model) and -3% (LLNL model). There are many possibilities on how this model-derived ozone spread could arise, for example: 1) the accuracy in the assumptions made in the chemical solution approach (e.g., an explicit numerical solver vs. an implicit Gear solution approach); 2) difficulties in representation of lumping of chemical species into groups or families; 3) different dynamical representations and the impact of these representations on ambient species distributions.

This study *will not* attempt to determine which of the five modeling approaches may be most accurate, instead we present preliminary results with the goal of understanding the uncertainties in the published LLNL results only.

All previous versions of the LLNL 2-D model have been integrated in a diurnal average manner, using diurnal averaging coefficients derived off-line from a fully diurnal model. The diurnal model was integrated for single days at eight seasonal times, producing diurnal averaging coefficients

at each altitude and latitude grid zone, for both thermal and photolyic reactions. In the diurnal average model execution, coefficient values between these seasonal times were interpolated. In this model, the continuity equation was solved for each chemical species (no chemical lumping approximations were made) using a Gear numerical solution approach. The potential error in this chemical solution approach lies in the assumptions made in deriving and using diurnal average coefficients. In order to evaluate the accuracy of the published LLNL HSCT assessment, a new 2-D model of the troposphere and stratosphere was developed.

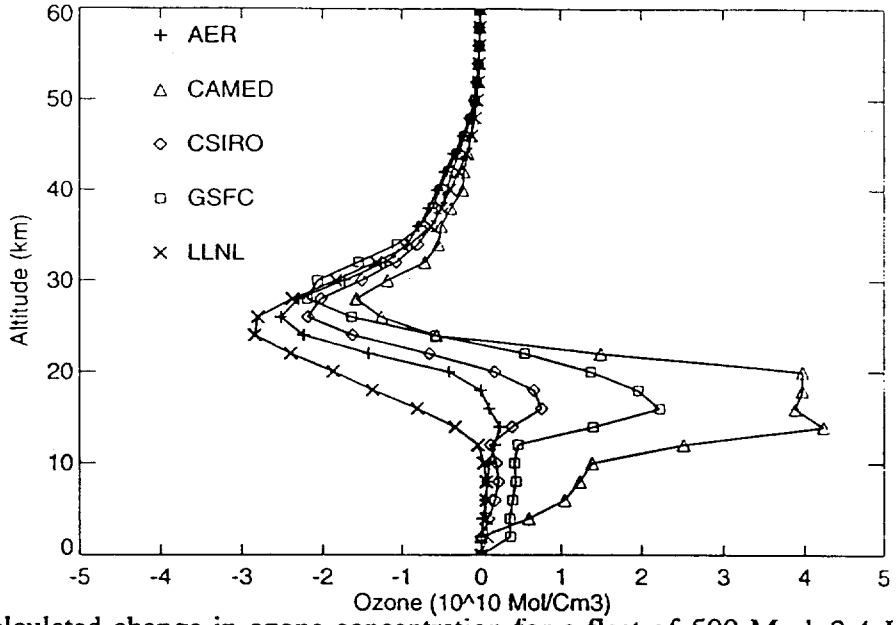

Figure 1 — Calculated change in ozone concentration for a fleet of 500 Mach 2.4 HSCTs (EI_{NO_x} = 5). The results are shown at 45°N for the month of March. This plot is taken from the NASA 1995 Scientific Assessment of the Atmospheric Effects of Stratospheric Aircraft, Figure 24 [1].

2. LLNL 2-D MODEL (*LOTUS*)

The new LLNL 2-D chemical-radiative-transport (CRT) model or *LOTUS* (*L*ivermore *O*perator-split *T*wo-dimensional Zonal-average (*U*) *S*ystem), determines the atmospheric distributions of chemically active atmospheric trace constituents in the troposphere and stratosphere. The individual components (i.e., chemistry, advection, diffusion) from the previous LLNL 2-D CRT [2] have been improved and modularized and are solved in an operator split manner. LOTUS can be integrated in either a *diurnal* or *diurnal average* manner. The model domain extends from pole to pole, and from the surface to 84 km. The horizontal resolution is 5 degrees in latitude and the vertical coordinate corresponds to the logarithm of pressure, with a resolution of 1.5 km. Below is a brief description of the photochemical component of LOTUS — for a description of the dynamical and radiative processes see Kinnison et al., 1994 [2].

The photochemistry represents the tropospheric and stratospheric interactions of actinic solar flux and the species families O_x, NO_y, ClO_y, HO_y, BrO_y, and CH_4 and its oxidation products. The chemical mechanism incorporates 48 transported species. There are 101 thermal and 44 photolytic reactions. Source gases present in the model include NO_x, N_2O, CH_4, CO_2, CO, the chlorine containing compounds CFC-11, -12, -113, -114, -115, HCFC-22, CCl_4, CH_3CCl_3, CH_3Cl, and the bromine containing compounds CH_3Br, CF_2ClBr, and CF_3Br. Most of the thermal reaction rate constants were taken from the NASA Panel recommendations provided in JPL Publication 94-26 [3]. Absorption cross section information was assembled from a variety of sources, including JPL 94-26. Hydrolysis of $ClONO_2$, N_2O_5, and $BrONO_2$ on the surface of stratospheric sulfuric acid aerosol are included as the probable dominant heterogeneous process. Neither N_2O_5 nor $BrONO_2$ have a sulfate aerosol composition dependency; their reaction probabilities are 0.1 and 0.6 respectively. $ClONO_2$

hydrolysis does have a composition dependency and is modeled following the work of Hanson et al. [4]. This study does not include any representation of chlorine activation by polar heterogeneous chemistry. In the photochemical operator, the continuity equation is solved for *each individual species* (i.e., no lumping of species into chemical families are made) using a variable time step, variable order, implicit technique for solving stiff numerical systems with strict error control. This solution technique (SMVGEARII) has recently been developed by Jacobson [5].

All model results from this study are consistent with integrating the continuity equation in a *diurnal manner*, therefore eliminating any errors incurred by integrating in a diurnal average mode using diurnal average coefficients.

3. LOWER STRATOSPHERE ODD-OXYGEN LOSS

Before any model is used in an assessment calculation, it should be evaluated when possible with observations. Within the last few years the amount of data available for model/data intercomparisons has increased dramatically with the launch of Upper Atmospheric Research Satellite (UARS) and the numerous ER2 aircraft campaigns. Below is a example of how results from one aircraft campaign can be used to evaluate lower stratospheric ozone photochemistry.

Aircraft measurements of simultaneous trace constituents distributions during the Stratospheric Photochemistry, Aerosol and Dynamics Expedition (SPADE), allow one to determine the odd-oxygen loss partitioning among chemical families [6]. This data was taken in April and May 1993, between 15-60°N, up to 21 km. The sulfate aerosol surface area density (SAD) was about five times the expected volcanic clean background. As discussed in Wennberg et al. [6], this data highlights the dominance of HO_x and ClO_x+BrO_x catalytic loss relative to NO_x during this period. Comparison of this data to model-derived partitioning is an important test of any assessment model. In *Figure 2*, comparison of a LOTUS result to the SPADE data is shown. Here the ambient atmosphere of LOTUS is representative of a 1993 atmosphere with five times the background SAD. The comparison between data and model-derived odd-oxygen loss is in good agreement for the latitude shown. However, more detailed analysis of not only the odd-oxygen partitioning, but the absolute NO_x, HO_x, and ClO_x+BrO_x odd-oxygen loss rates is needed before one develops trust in the accuracy of a model representation of ambient conditions.

4. HSCT ASSESSMENT RESULTS

As shown in *Figure 1*, the LLNL 2-D model result published in the 1995 assessment [1] is considerably more negative in the lower stratosphere (less than 20 km) than the other four assessment models. After development of LOTUS and comparison to available data, we have re-integrated a few of the 1995 HSCT assessment scenarios to see what effect our previous use of diurnal average coefficients had on our published results. In addition to changes in our diurnal average approach, there were additional improvements to LOTUS over the previous LLNL 2-D model used in the 1995 assessment. These differences include: 1) re-evaluation of tropospheric lightning, increasing the integrated source from two to five Tg $year^{-1}$; 2) calibrating the methlychloroform lifetime to 5.0 years (previous 6.9 years); 3) lowering the location of the minimum in the water mixing ratio within the lower stratosphere by up to two grid zones; 4) including $BrONO_2$ hydrolysis with a reaction probability of 0.6; and 4) assorted computational "bugs". These individual difference will not be discussed in detail here, however, the sum of all these differences explain the majority of the difference between the new LOTUS assessment and that published in the 1995 assessment.

In *Figure 3*, the new result using LOTUS (EI_{NOx} = 5, 1xSAD; solid line) is shown and can be compared directly to *Figure 1*. The change in ozone is even more negative than previous published results. Comparison of percentage change in column ozone is shown in both *Table 1* and *Figure 4a* and *Figure 4c*. Here again, the LOTUS model-derived change in column ozone is more negative that previously published results. This increased ozone depletion is a combination of many factors, but

one can certainly say that the difference between the LLNL model and the other models in the 1995 HSCT assessment is *not due* to inherent errors in the previous LLNL diurnal averaging approach.

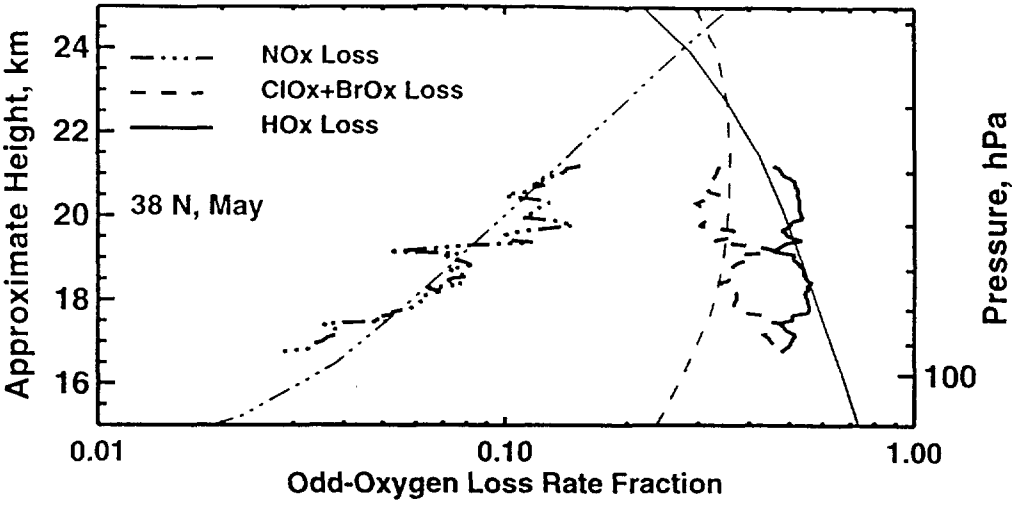

Figure 2 — Model-derived HO_x, NO_x, ClO_x+BrO_x, odd-oxygen loss partitioning (narrow lines) relative to data retrieved on the 1 May 1993 flight of the ER2 aircraft during the SPADE campaign (thick lines).

5. SURFACE AREA DENSITY SENSITIVITY

The magnitude of the sulfate aerosol surface area density has a large impact on the magnitude of an HSCT ozone assessment. In *Figure 3*, *Figure 4b*, *Figure 4d* and *Table 1*, the impact of increasing the SAD by a factor of five is shown. Local ozone change in the troposphere and lower stratosphere is positive. In the stratosphere, this is due to a repartitioning of odd-oxygen loss away from NO_x and towards HO_x and ClO_x+BrO_x. In the troposphere, more ozone is formed via the methane smog reactions. The net extent of both these processes is a less negative column ozone change. In future assessments it will be very important to examine how assessment model compare when different magnitudes of SAD are used.

Table—1 Calculated steady-state change in column ozone (%). The background case includes emissions from a projected 2015 subsonic fleet without HSCTs. The 1xBkg SAD is taken from [7] and was used in the NASA 1995 assessment [1]. The previous LLNL 2-D assessment (LLNL [1]) did not include $BrONO_2$ hydrolysis on sulfate aerosol.

Model	EI NOx	SAD	Global	N.H.	S.H.	40-50N
LLNL in [1]	5	1 x Bkg	-0.2	-0.3	-0.2	-0.3
LOTUS	5	1 x Bkg	-0.4	-0.4	-0.3	-0.5
LOTUS	5	5 x Bkg	-0.2	-0.2	-0.2	-0.2
LLNL in [1]	15	1 x Bkg	-0.7	-0.9	-0.4	-1.0
LOTUS	15	1 x Bkg	-0.9	-1.2	-0.6	-1.4
LOTUS	15	5 x Bkg	-0.1	-0.2	-0.1	-0.1

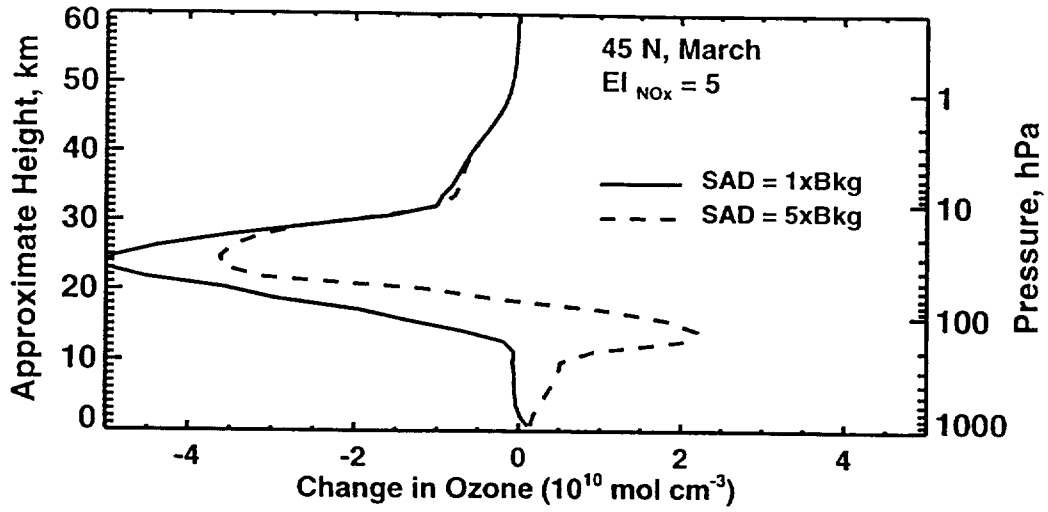

Figure 3 — Calculated change in ozone concentration for a fleet of 500 Mach 2.4 HSCTs (EI_{NOx} = 5).

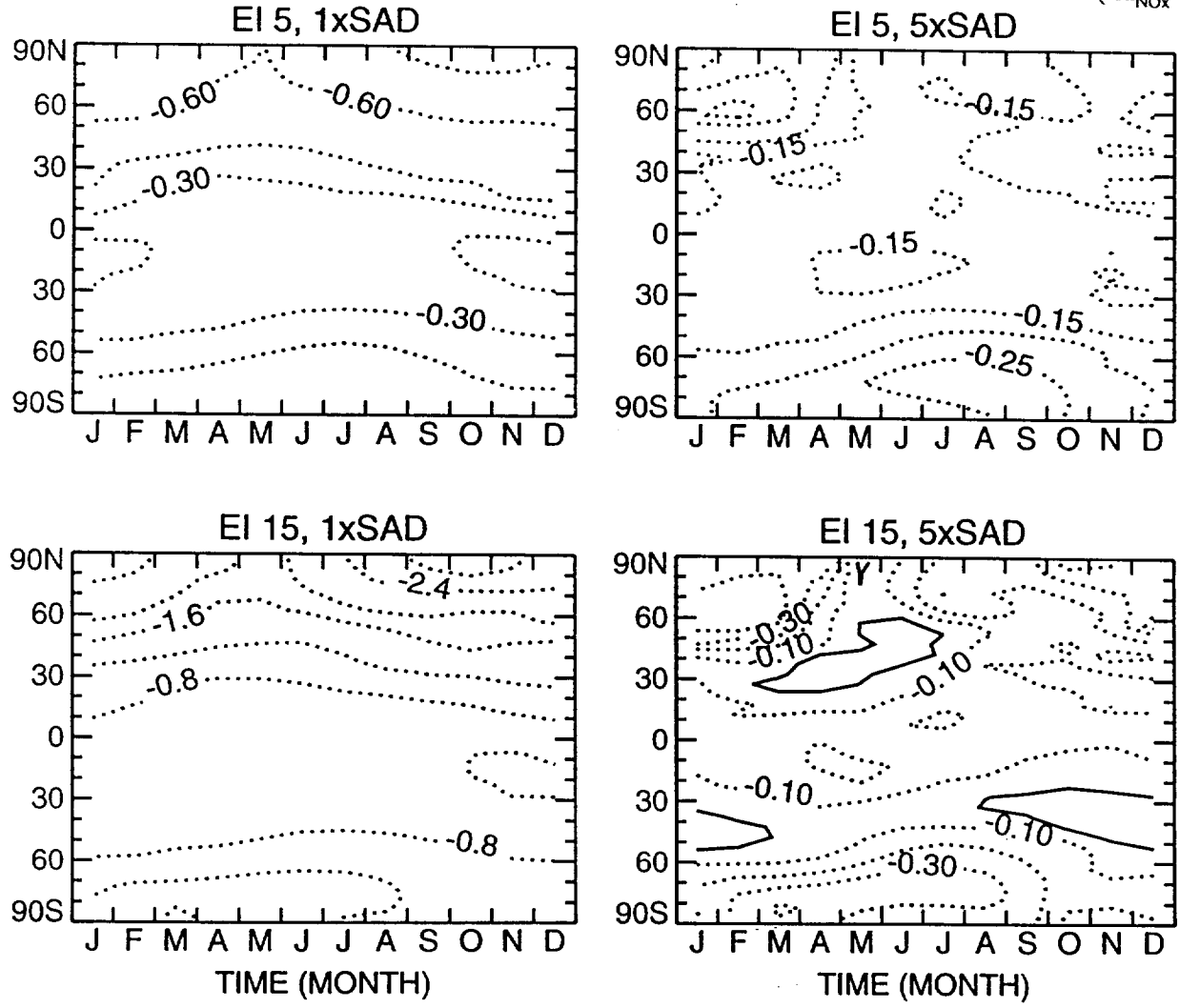

Figure 4 — Percentage change in column ozone: a) EI_{NOx}=5, SAD=Background; b) EI_{NOx}=5, SAD=5xBkg; c) EI_{NOx}=15, SAD = Bkg; and d) EI_{NOx}=15, SAD=5xBkg.

5. SUMMARY

The new LLNL 2-D model (LOTUS) was used to investigate the impact of emissions from HSCTs on stratospheric ozone abundance. This model is integrated in a diurnal manner (diurnal average coefficients are not used), making no chemical family lumping assumptions. Initial comparison to aircraft data suggest good agreement in odd-oxygen loss partitioning. More detailed comparison of absolute ozone loss is still necessary before definitive conclusions on the agreement between data and LOTUS can be made. Proposed HSCT emission scenarios using LOTUS gave a more negative ozone response than the previous LLNL 2-D model and other 2-D assessment models. The sensitivity to sulfate aerosol SAD is large and should be included in the next international assessment of the atmospheric effects on stratospheric aircraft.

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